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EFFECT OF IRRADIATION AS A
MATERIAL SCIENCE PROBLEM

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The importance of physical metallurgy or "Material science" for nuclear engineering hardly needs special emphasis. This particularly applies to that part of physical metallurgy which is concerned with the effect of irradiation on materials.

It is evident that without knowing changes in structure and properties of materials suffering "radiation damage" one can neither design nuclear reactors reasonably nor make reliable predictions concerning their operation. In many cases the accumulated metallurgical experience cannot be taken advantage of to choose reactor materials first of all because many materials operate for the first time and secondly because in this case engineering faces a very composite combination of operating conditions and requirements of which metallurgy was not aware of earlier.

The novelty and difficulty in understanding changes occurring in materials under irradiation call for their most careful study and first and foremost for the study of fundamental phenomena of irradiation effects.

For a really reliable prediction it is not sufficient to be confined to a simple statement of experimental data, no matter how extensive they may be. It is necessary to develop a stringent and comprehensive theory as deductive as possible the faith in the conclusions of which would be obligatory not only for theoretical science but also for a practical designer.

However, the objectives of radiation material science are wider than merely assisting a nuclear engineer in his choice of reactor materials. It opens up the way to the field of basic problems of solid state physics. Giving a method of a basically controlled introduction of defects and imperfections of various kinds into a material, the irradiation should play the main part in understanding those properties of a solid body which depend upon defects of a crystal structure, in other words, in understanding almost all the properties of a real crystal. So far as the mechanics of solid bodies is concerned, such phenomena as strengthening, relaxation and creep, intrinsic friction, ductile and brittle fractures belong here. So far as physico-chemical phenomena are concerned radiation effects may throw light on many problems of both equilibrium and kinetics of phase transformations, ageing, ordering, diffusion etc. A study of an interaction of fast particles with a substance should greatly advance the science dealing with the strength of materials and eventually contribute to a development of those "supermaterials" in the absence of which the modern technological progress will remain rather limited.

Finally, as a third link that can connect radiation material science with practical needs, one may also mention a direct use of a radiation exposure to improve useful properties of materials. At present one can hardly do more than attempt to imagine the future development of this branch of technical science since a fairly great progress should be made in designing appropriate irradiation facilities and solving a great number of other questions before it becomes possible to speak about this task as being feasible.

To my mind, the former of the mentioned above aspects interests the most part of the participants of of the colloquim to a lesser extent; the third one is more of nature of a desire rather than of an achievement, that is why I expect to have more opportunity of drawing your attention if I deal with the second one, i.e., with the importance of studying radiation defects for the general progress in our knowledge of material science.

Changes in a solid structure induced by energetic particles of neutrons and fission fragments may be divided into two groups a)Those characteristic of a solid material under normal conditions as well and produced by common means such as plastic deformation, heating, low impurity alloying etc and b)Those not encountered generally, however, being the result of applying the utmost force best reveal the ability of the crystal medium of a solid material to withstand any trial to disturb the lattice and the ability to restore it. Those exaggerated effects show us as well upon what imperfections and to what extent the properties of a solid material depend.

Two examples will be given below as to the radiation effects of both the first and the second kind. The investigations by Bohr, Seitz, Dienes, Cottrell, Drink-

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man and oth. have lead to the general conclusion that the collisions of fast neutrons with atoms of some substance result in the appearance of highly mobile displaced atoms and somewhat less mobile vacancies in a crystal lattice. Under ordinary conditions of irradiation the concentration of these defects rarely exceeds 1-2%. If the temperature of a substance being irradiated is not very low, a considerable part of these defects disappears eitherdue to mutual annihilation or by combining into clusters or in some other way, e.g., by trapping defects on imperfections existing in a crystal lattice. While migrating defects can give rise to many transfer phenomena (diffusion, elastic stress relaxation, order-disorder transitions etc) which can result in the substance both approacing physico-chemical equilibrium state and departing from it. The formation of clustered interstitials and vacancies interferes seriously with the movement of dislocations, increasing their friction when they move and , consequently,strengthening a material.

Together with the notion about producing single defects (Frenkel defects) by a collision of a fast bombarding particle- neutron- with individual atoms in its path and by successive pair collisions of a primary knocked on atom with the other atoms of a lattice there existed a different notion about the group formation of a great number of defects, by a fast atomic particle (primary knock-ons, slowed down fission fragments, energetic ions from some other source) exchanging its energy simultaneously with a large number of other atoms in a restricted volume so that a phenomenon similar to a burst occurs.

The atoms of this region fly apart leaving behind a void in its centre at its periphery excessive atoms accumulate and accomodate in some way or other between the atoms of a medium. Such a state being most unstable may last only for fairly short time (10^{-12}-10^{11} sec). Following it the partial recovering of the damages occurred may take two paths (Brinkman [10]). Either at the next moment a void, formed in the centre of the region, will be filled with atoms returning from the periphery, this will result in the restoration of a damaged lattice. In this case the atoms if one compares their positions with the initial ones, will be accomodated on some other lattice sites(here according to Brinkman

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the term "displacement spike" comes). Or the atoms, knocked out to the periphery, will not be able to return in the centre on account of a rapid loss of their energy due to dynamic collisions and the remaining cavity will either exist in the form of a bubble or will collapse forming a dislocation loop. In this case the displaced atoms being very mobile may easily produce clusters in the form of interstitial loops.

No matter in what way will consequences of a burst be healed, the atoms of this region will preserve a considerable excess of the kinetic energy for some time. If the rate of an energy removal from a "spike" region is not sufficiently high and during perturbation atoms will have time to make a great number of oscillations, this region may be considered to be in a state of intense heating. However, it appeares that in a number of cases it may be suggested that the energy leaves the place of slowing down rapidly transferring an impulse from one atom to another in a dynamical fashion by successive atomic replacements similar to those shown by Gibson and Vineyard for a low(< 400 ev) energy of a primary particle [9]. That is why, the question of the validity of the ther thermal interpretation of a "displacement spike" and

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the related question as to the occurrence of radiation annealing are not quite clear. In every case the occurrence of radiation annealing should be proved experimentally. The total amount of the released energy is likely to serve as a determining criterion here.

One of the examples of a phenomenon in which the main part is played by single mobile defects produced by neutron bombardment is an experiment on the relaxation of elastic stresses. The description of the experimental part has been given earlier [2-4]. The same phenomenon takes place in springs of various nonfissile materials. Here one can observe a similar relaxation phenomenon too it is rather slower than that in fissile materials but fairly appreciable in some materials such as nickel. A relaxation phenomenon is also observed for microstresses, the amount of which is easily controlled by the width and the form of x-ray lines recorded by a diffractometer.

An essential feature of a radiation enhanced relaxation is rather a considerable effect of reversion on tempering at an ordinary temperature or on slight heating a specimen, that suffered a radiation induced relaxation. The springs show a tendecy towards an inverse deformation, i.e. a change of the deflection of the springs

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and in case of inner microstresses this reversion results in the secondary broadening of the x-ray lines that were narrowed on the relaxation.

In all the cases the relaxation phenomenon is the fuller the less the amount of the previous irradiation. It should be pointed out that a) previously irradiated materials also have the capacity for accelerated relaxation, b) for materials of high crystallization temperature such as ,an additional slight heating is required to produce a noticeable relaxation effect after irradiation. c) a relaxation effect is greatly decreased with lowering the temperature of a specimen being irradiated (-70°C).

All the above mentioned facts can be easily accounted for by the following scheme. Irradiation produces highly mobile defects which move towards stress concentration regions under the action of an elastic field gradient. Since interstitials and vacancies make different contributions to the average atomic volume, the defects of both these types shift in the opposite directions leading to the decreasing of residual stresses. At this stage the defects are still sufficiently mobile and on slight heating can leave "traps" and

and annihilate. It is in this respect that the phenomenon of reversion is likely to consist. At the next stage with an increase in the local concentration of the defects of one sign their condensation occurs which may be accompanied by the formation of dislocation loops. The decrease of the reversion effect on prolonging the irradiation exposure may be associated with the segregation of the condensed defects.

The temperature dependence of the relaxation is indicative of the fact that the thermal activation is necessary for the radiation induced defects to be shifted in the field of stresses. It should be noted that in general the behaviour of a fissile material (U) is similar to that of a non-fissile one: the difference being only in the rate of a process. That is why the mechanism in both the cases should be considered to be essentially the same - migration of single defects to stress concentration regions. (In fissile materials this may be accompanied by the preferred survival of the defects of one sign corresponding to the sign of deformation [11]).

If such a model is true, then the comparison of the rates of the relaxation process in fissile and non-fissile materials shows that the number of the irradiation

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induced defects related to the number of fission events (of scattering respectively) in fissile materials (natural uranium) should be 100- 1000 times more than in non-fissile ones.

The experiments on the homogenization of some fissile alloys show an example of another type of the irradiation action on materials when the results may be interpreted as being the consequence of radiation annealing. As previously established [1,2] ,the neutron irradiation of a U + 20at% Mo alloy induces the transformation of a eutectoid, consisting of α-uranium and an intermediate compound U_2Mo , into a γ-base homogeneous solid solution. The theory accounts for this phenomenon by the fact that in the regions of slowing down fission fragments, having the approximate size of $10^{-17} cm^3$, a complete atomic mixing takes place on account of heating to high temperatures. The successive overlapping of such regions of heating should result in equalizing an average concentration, which is equivalent to a forced radiation diffusion.The conclusion drawn from the theory to the effect that the rate of homogenization should depend on the square of the linear dimensions of a dispersed phase is confirmed in work [5,2] .

..e fact that it is the radiation annealing produced by fission fragments and not some specific properties of α-uranium matrix, that is responsible for this phenomenon was confirmed in a later study dealing with a similar process of homogenization of irradiated Cu-Sn bronze containing a small addition of Pu (1at%) [6].

The low-temperature annealing (220°C) of an 8.2at% Sn - 1at% Pu - Cu alloy results in a highly dispersed precipitate from an α-solid solution of an intermetallic compound Cu_3Sn (ε-phase); 3.1at% Sn remaining in a solid solution. After irradiation in the "..FT" reactor the amount of an intermetallic compound Cu_3Sn is decreased, and the saturation of the solid solution by Sn is increased attaining 5.5at% Sn after the irradiation in 6×10^{19}nvt.

An increase of an α-solid solution lattice parameter which is indicative of the growth of Sn concentration is well observed after irradiation by a flux of no more than 1.5×10^{18}nvt. The theory of the radiation diffusion previously developed for a U+20at%Mo alloy may be applied to the present case as well. It shows that the homogenization phenomenon of 8at% Sn - 1at% Pu - bronze is well accounted for on the assumption that the thermal spikes created by plutonium fission fragments produce radiation annealing on account of which an intermetallic compound is dissolved and tin diffuses into the solid solution. The calculated amount of the energy dissipated by a fission fragment in a thermal spike proves to be equal to that obtained in the case of irradiating the U-Mo alloy.

Although the homogenization of bronze with 1at%Pu added proceeds most effectively, the irradiation of a binary alloy Cu - 8at%Sn without plutonium by a considerable fast flux ($5x10^{19}$nvt) does not give rise to any appreciable transformations in it. If in accordance with the previous data one assumes that about 2 Mev of the energy released in the thermal spikes fall per one fission event and consideres the average energy of a primary knock-on on collision with 1 Mev neutron to be about 30 Kev, then the total effect of a heat release of irradiation annealing in a fissile alloy for a flux of 10^{18}nvt should be only two times more than in a non-fissile one (Cu - 8at% Sn) at a fast flux of $5x10^{19}$.

The absence of traces of the homogenization effect in the latter case questions the occurrence of radiation annealing in non-fissile materials bombarded by neutrons altogether.

However, the diffusion effects depend not only on the total amount of the energy released during radiation annealing but also on the square of the linear dimensions of an annealing region and owing to this the effect in non-fissile materials should be less. Therefore for the final solution of this question the flux to irradiate the binary bronze should be increased by at least an order of magnitude.

It is interesting to note that the Cu-Sn bronze being not susceptible to the displacement spike effects seems to be most susceptible to the phenomena associated with the existence of single radiation defects. The broad x-ray lines of the cold worked Cu - 8at% Sn alloy show considerable narrowing after irradiation. This is indicative of the fact that the mobile defects capable of inducing micro-stress relaxation are produced and accumulated in this alloy under fast neutron irradiation.

After all that was said concerning the influence of irradiation on metals a question arises that could become a subject to discuss at the present colloquim. Let us formulate it in the following way.

It is known that at a low initial energy transferred by a fast moving particle to an atom, the latter dissipates it to transfer impulses to neighbouring atoms. As it was assumed by Silsby [9] and proved by Gibson and oth. [8] in their experiments with a computer the energy transferred is primarily concentrated in certain directions of a crystal lattice and used giving rise to a number of successive atomic replacements along the chain. Such a propagation of the energy is of a "dynamic" nature. atomic oscillations in the chain rapidly attenuate, on account of which all the replacements occurred as well as the displacements at the end of the chains and on their branches "freeze" rapidly. Such a dynamic or "cold" damage may be distinguished from a "hot" or statistical one. The latter will take place if the transferred energy is so high that its propagation will occur not only by the transfer of the impulses to the neighbouring atoms sitting firmly in the lattice sites or displaced by one or two atomic distances only but also by a simultaneous displacement of many atoms having large kinetic energy. In other words, if the propagation of an impulse along the atomic chain is called a "focusson"(the term accepted lately) then in case of a hot damage the movement of each atom is a result of an interference of many focussons, issuing from many centres.

It will be quite natural to assume that the criterion determining the type of a damage must be looked for in an amount of the energy with which slowed down particle arrives at a region where its path length is comparable with atomic spacings.

What is the value of the energy and where should a boundary line be drawn, however?

The question raised is reduced to this.

Now one more experiment concerned with the influence of irradiation will be described. It refers to the question raised. By the way, it can serve to illustrate how the influence of irradiation on materials helps better understanding the nature of the phenomena occurring in materials under ordinary conditions.

As established from the many earlier experiments irradiation of ordered solid solutions results in their rapid disordering. In fissile materials it is undoubtedly due to the thermal spike effect produced by fission fragments. It may be seen from the fact that the disordering process is completed for the time equal to that necessary for the whole metal volume to pass through the thermal spike state. However, the disordering mechanism in non-fissile materials is less clear.

Here it may be the consequence of both the dynamic and statistical damage.

G.P.Saenko investigated a Fe-25at%Al alloy. On slow cooling from 580°C the alloy turns out to be ordered, however, after quenching in oil from 750°C it becomes partially ordered retaining a short range order in the absence of a long range order.

The peculiar feature of this alloy is the fact, that such partial ordering is characterized by increased electrical resistance. Residual ordering of a quenched alloy can be destroyed by plastic deformation, along with this its electrical resistance is decreased.

Irradiation brings about a considerable growth of $\Delta R/R$ in an annealed alloy and some decrease of its resistance in a quenched one. After irradiation by a flux of 1.5×10^{20} n/cm^2 both the specimens have practically the same resistivity value. This as well as the x-ray diffraction data point to the fact that both the alloys are in a similar condition of almost complete disordering.

The behaviour of cold worked alloys is of interest. In this case the electrical resistance is drastically increased at low fluxes, which seems to be indicative of the restoration of the short range order destroyed by deformation;however, after a slight decrease the electrical resistance remains practically unchanged.

The behaviour of irradiated alloys differs characteristically on subsequent annealing.

Matters stand as if both the alloys kept memory of their initial state. This seems to suggest the "dynamic" mechanism of the disordering process under irradiation. Some quantitative calculations bring us to the same conclusion.

If one considers the disordering of an annealed alloy to occur through the action of displacement spikes, then from the value of the flux required for almost complete disordering ($2x10^{20}$nvt) it is possible to estimate the size of a displacement spike volume on the assumption that complete disordering is achieved in the "spike" volume. If one takes $3x10^{24}cm^2$ as the value for a scattering cross-section then the number of primary knock-ons in 1cm^3, necessary for disodering, will be $0.5x10^{20}$ and the volume of one spike would be $2x10^{-20}cm^3$.

Then on the basis of the data of work [5] one can easily deduce that the energy stored inside such a volume will hardly be more than 2000ev, i.e., it will not exceed approximately 1/20 of the average energy transferred by a fast neutron of 1 Mev to a primary atom colliding with the neutron. Thus, if the mechanism of the "hot" damage or radiation annealing plays some part in the disordering of a Fe-Al alloy, this role is not great as compared with that played by damages, produced in dynamic processes of atomic collisions

Conclusions

The following conclusions may be drawn from the above said:

1. The study of the effect of irradiation on metals and alloys should undoubtedly contribute to better understanding the nature of the phenomena taking place in a great number of metallurgical processes and , consequently, make for an intensified control of these processes.

2. To fully use the information obtained in investigations of the influence of neutron irradiation on materials, it is necessary first of all to solve some problems concerning the principal results of this effect. Primarily it is necessary to distinguish between the phenomena in which the main part is played by single,

mobile defects produced by bombarding particles and the phenomena associated with larger damages that may be thought of as equivalent to a release of a fairly great amount of the thermal energy in a restricted volume.

3. The report shows that the radiation induced relaxation of elastic stresses in both fissile and non-fissile pure metals and alloys is a typical example of a phenomenonon in which the main part is played by single defects then binding together in clusters.

4. The radiation induced homogenization of the alloys, containing dispersed precipitates, may serve as an example of the second type phenomena, when a determining factor proves to be irradiation annealing taking place in a thermal spike region. The role of a fissile addition (Pu) is shown by an example of the homogenisation of Sn-bronze, an alloy having the composition of Cu-8.2at%Sn-1at%Pu. Alloys without this addition do not show any transformation effect although single defects seem to accumulate in this material, this is revealed by a considerable elastic stress relaxation phenomena observed in the cold-worked Cu-Sn alloy under fast neutron irradiation.

5. The experiments are discussed as to the effect of neutron irradiation on ordering Fe_3Al alloy. The

disordering of the ordered solid solutions has previously been considered either to be an effect of displacement spikes or to be a consequence of replacement collisions. In this case the study of the rate of a disordering process in the annealed Fe_3Al alloy as well as a "memory" phenomenon on annealing irradiated alloys, initially taken both in the short range and long range orders is more in favour of the "cold" type of a damage, i.e., of the fact that under irradiation the disturbance of an order results from the chains of collisions or from the so-called "focussons".

6. When should one expect one or the other type of a damage, dynamic (cold) or statistical (hot) and on what parametres (associated either with a bombarding particle or with a medium) does it depend - are questions which require further theoretical and experimental investigations.

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